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Photon Echo Spectroscopy in the Single Optical-Cycle Regime

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10.00 CMB6

Laser written waveguides in glasses and crystals

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Waveguides for integrated optical applications are usually made by indiffusion of ions into crystal or glass materials. However, the generation of three-dimensional structures is hard to achieve. This problem can be overcome by laser generated waveguides.

Ultrashort laser pulses focused inside optical transparent bulk materials create local structural changes and in consequence of this a refractive index gradient. The result of moving the sample through the laser beam is a refractive index profile like in a buried waveguide.

This direct writing method offers the potential for three-dimensional waveguide structures and allows integrated optics in different materials.

The experimental setup is shown in Fig. 1. The laser pulses that we used to induce refractive index changes were obtained from a regeneratively amplified $\text{Ti}^{3+}:\text{Al}_2\text{O}_3$ laser. The duration of the 780 nm pulse was 120 fs and the repetition rate of the laser was 1 kHz. The laser beam with a pulse energy of a few microjoules was focused into the material. The sample was fixed on a computer controlled XYZ stage.

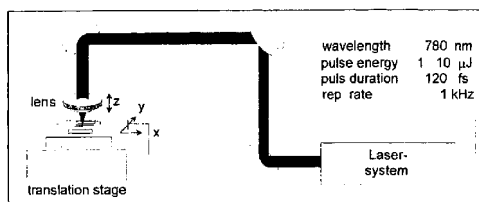


Fig. 1

Generation of waveguides in fused and crystal silica is presented. Properties of the waveguides are discussed.

10.15 CMB7

Photon Echo Spectroscopy in the Single Optical-Cycle Regime

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A very high temporal resolution and a broad bandwidth are but two advantages provided by the use of extremely short sub-5-fs pulses [1] in a nonlinear spectroscopic experiment. However, the applicability of the standard theoretical description becomes questionable for the pulses that consist merely of a couple of optical oscillations. For instance, the conventionally employed slowly varying envelope approximation, implying that the change of the pulse amplitude on the duration of an optical cycle is negligible compared to the magnitude of the amplitude itself, can no longer be maintained. Furthermore, the phase-matching bandwidth that is limited due to dispersion in the nonlinear medium rapidly gains importance with the increase of the spectral width of the pulse. Another point of serious concern is the frequency-dependent variation in the sensitivity of signal photodetectors. In combination, the above listed features result in what is known as a spectral-filter effect. Finally, artificial lengthening of the experimental transients is a direct consequence of the noncollinear geometry employed in spectroscopic experiments.

In this Contribution we present a theoretical analysis which thoroughly reexamines the formalism of ultrafast photon echo spectroscopy. We obtain a general expression for the echo signal, which is valid even for single-cycle-pulse applications. The derived formalism is applied to photon-echo spectroscopy on the hydrated electron with 5-fs pulses.

According to our calculations, a careful choice of the beam geometry and selection of a photodetector with the suitable spectral sensitivity defeats the otherwise damaging role of the spectral-filter effect as well as that of geometrical smearing. Importantly for the weak-signal applications such as photon echo spectroscopy, the absence of spectral filtering eliminates the otherwise unavoidable requirement to frequency-resolve the signals.

Figure 1 presents the two-pulse photon echo signals obtained from the neat water and electrons, solvated in water. A minute difference in the widths of these two traces suggests that the electronic dephasing of the hydrated electrons is extremely fast. The finite population lifetime of the electrons in the excited state causes the delay of the echo trace in Fig. 2b. The best fit to the experimental data yields a pure dephasing time of $T_2 = 1.6$ fs. With this value, the absorption spectrum of the hydrated electron can be successfully modeled provided the conventionally used rotating wave approximation is abandoned [2].

References

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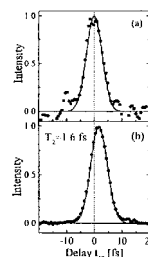


Fig. 2. Results of two-pulse photon echo experiments on water alone (a) and hydrated electrons (b). Circles represent experimental data points and solid curves show results of simulations.